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Progress of the Phase-change Optical Disk Memory

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1. Introduction

Optical memory has two recording modes: the photon-mode as a silver halide photograph and the heat-mode as a laser optical disk. Though laser heat-mode recording has the advantage of environmental stability, it has limitations due to thermal diffusion phenomena, which will be discussed in this paper.

Optical disk memory has the unique feature of read-only media performance, which is also compatible with the rewritable function and is different from HDD (hard disk drive) technology.

Rewritable optical disk technology progressed with the race between the magneto-optical (MO) disks and the phase-change rewritable (PCR) optical disks. With the increasing use of multimedia, phase-change rewritable optical disks are becoming more popular due to their CD (compact disk) and DVD (digital versatile disk) compatibility.

In 1968, S. R. Ovshinsky discovered a new memory phenomenon in chalcogenide film materials. This order-disorder phase-change memory effect came to be called the "Ovonic Memory"⁽¹⁾. In developing this storage medium, the main issues have been the stability of the film materials, the stability of the reversible cycle characteristics and the recording sensitivity. The author and his colleagues were the first to achieve a breakthrough in these areas, which led to the commercialization of phase-change optical disk products. The first version of the phase-change optical disk product was shipped in 1990 from Matsushita/Panasonic. The PD(phase-change dual) and CD-RW(rewritable) followed, and now a rewritable DVD with 4.7 GB capacity and 3.4 Gbit/in² density is being produced.

Blue laser technology, large numerical aperture lens, volumetric recording and multi-level recording technologies are candidates for the future of high-density phase-change recording technology.

This paper describes the phase-change optical disk memory progress of high-density recording, phase-change optical disks with a density of approximately 100 Gbits/in² and more, and discusses the basic effect of the ultra-short laser pulse of the femto second laser pulse response on phase-change media.

2. Principle of the phase-change overwrite memory

2.1 Overwritable phase-change material

The rewritable optical memory phenomenon has been observed in $\text{Te}_{81}\text{Ge}_{15}\text{Sb}_2\text{S}_2$ composition material⁽²⁾. This material was modified from a $\text{Te}_{85}\text{Ge}_{15}$ eutectic composition by adding Sb and S elements. Figure 1 shows the phase diagram of the Ge-Te system. At the eutectic composition, the melting temperature decreases to 375°C. In the early stage of the investigation of phase-change materials, it was important to obtain simple amorphizing compounds, and therefore the eutectic compound composition was chosen. For the eutectic composition, the melting temperature is at a minimum and viscosity is expected to increase. It is thus easy to freeze the bonding structure in the liquid phase through the cooling

process. In the next stage, applicable materials that have rather high-speed crystallization characteristics were used.

Phase-change materials for over-writing using one laser spot need to have high-speed crystallizing characteristics. Thus, high-speed crystallizing materials such as the In-Se system were discovered³⁾. Then Ge-Sb-Te system was proposed^{4, 5)} and in the GeTe-Sb₂Te₃-Sb system shown in Fig. 2, the material shows nucleation dominant crystallizing characteristics⁶⁾. Recently, at the ODS (optical data storage conference, Whistler, May 14-17, 2000) crystallizing growth-dominant compositions in the eutectic composition system, such as Sb₆₉Te₃₁, were proposed for high density and high data-rate recording⁷⁾.

2.2 Model of the phase-change memory

The phase-change processes of the Ge-Sb-Te amorphous material were examined by DSC (differential scanning calorimeter) measurement. Figure 3 shows that there are two exothermal peaks and one endothermal peak. The first exothermal peak corresponds to the crystallization phase-change and the second to the fcc-to-hexagonal crystalline structure change.

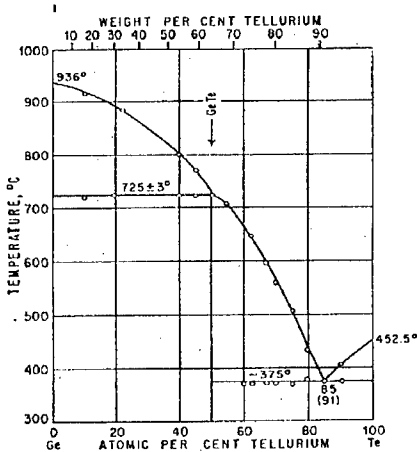


Fig.1. Phase diagram of Ge-Te system.

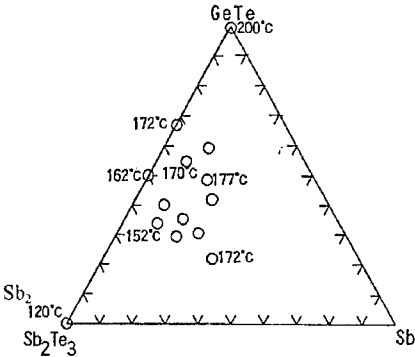


Fig. 2. Crystallizing temperatures of the GeTe-Te₃Sb alloy system.

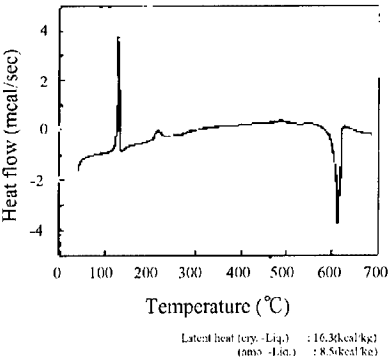


Fig. 3. DSC (differential scanning calorimeter) analysis of Ge-Sb-Te film at a heating rate of 10 °C/min).

The third peak, the endothermal peak, corresponds to the melt-phase transition⁸⁾. The latent heats of crystalline to liquid (16.3 kcal/kg) and amorphous to liquid (8.5kcal/kg) were obtained from this measurement, the later was calculated as the subtraction value of the exothermic heat from the former value. Figure 4 shows the model of the phase-change memory. The enthalpy of the amorphous state and the crystalline state is different. The complex refractive index $N = n + ik$ of the film is different for the two phases. When the cooling rate is above the critical cooling rate (3.4 K/ns) of amorphizing, the portion of the film at "f" enters the amorphous phase⁹⁾.

3. Key technologies of the phase-change optical disk media

3.1. Thermally-stable new dielectric protection layer

In the early phases of its development, the most important subject of the phase-change optical disk was cycle degradation. Figure 5 shows a high resolution TEM (transmission electron microscope) image of ZnS and the new ZnS-SiO₂ mixture protection films. The grain size of the ZnS-SiO₂ film is very small, at around 2 nm¹⁰⁾. The new ZnS-SiO₂ dielectric layer is thermally stable and does not show grain growth, even after annealing at 700°C (5 min). Grain growth in the ZnS layer was one reason the phase-change optical disks degraded after many rewrites. The first version of the phase-change optical disk product with a 4-layer structure produced a greater than 100,000 overwrite cycle performance.

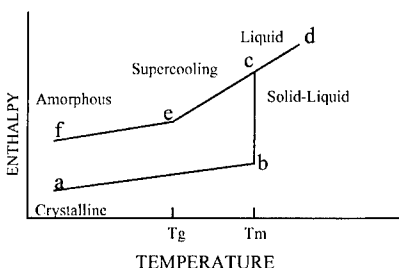


Fig. 4 Model of the phase-change memory

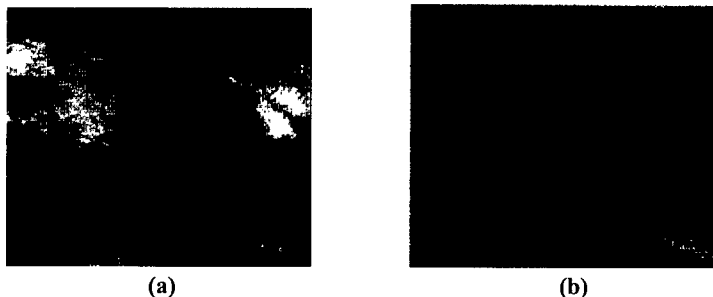


Fig. 5. High resolution TEM (Transmission electron microscope observation of ZnS (a) and new ZnS-SiO₂ mixture (b) dielectric films. layers.

3.2 Basic 4-layer disk structure and an additional SiO₂ layer

Figure 6 shows a cross-sectional TEM image of the basic 4-layer structure of a phase-change optical disk of PD. The layers comprise a protection layer, a bottom dielectric layer of ZnS-SiO₂ (155 nm), an active layer of GeTe-Sb₂Te₃-Sb (24 nm), an upper dielectric layer of ZnS-SiO₂ (45 nm) and a reflection layer of Al-alloy (100 nm). All the layers are sputter deposited on a polycarbonate disk substrate.

These layers work as a multi-layer optical interference structure, controlling disk reflectivity, which is the difference of the reflectivity between the amorphous mark and the crystalline erased state.

The other cycle degradation model is that the sub-nanometer level space deformation of the disk layers, which works as the motive force of the sub-nanometer displacement of the active layer components. The deformation occurs by thermal expansion of the layers along the thermal diffusion process. Table 1 shows the optical, thermal and mechanical properties of the disk layers.

Figure 7 shows the thermal simulation results of the temperature distribution in the cross-sectional view of the first version phase-change optical disk. The temperature distribution is calculated after the 30 ns laser-spot irradiation on the rotational disk. The linear velocity is $V = 8 \text{ m/s}$, the laser wavelength of is 830 nm and the lens numerical aperture is 0.53. It shows that the temperature rising area is wider than the spot size of $0.76 \text{ }\mu\text{m}$ (FWHM)(full width half maximum) caused by the thermal diffusion in 30 ns. The temperature distribution profile is asymmetrical between the forward area and the backward area because of the thermal diffusion process.

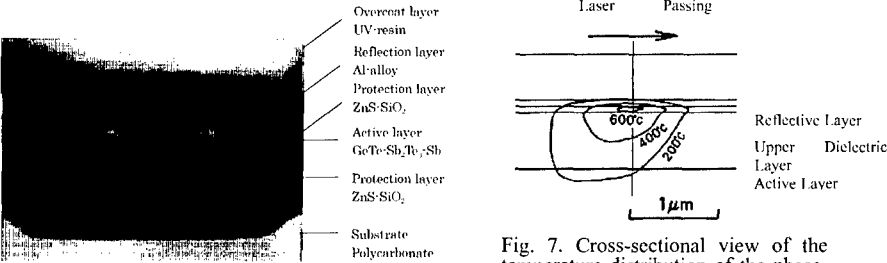


Fig. 6. Cross-sectional TEM observation of the basic 4-layer phase-change optical disk.

Fig. 7. Cross-sectional view of the temperature distribution of the phase-change optical disk at 30ns laser spot scan.

Table 1 Optical, Mechanical and thermal Properties of Materials

Material	Reflective Index $\lambda \sim 830\text{nm}$	Density (kg/m^3)	Young's Modulus (N/m^2)	Poisson's Ratio	Specific Heat ($\text{J}/(\text{kg}\cdot\text{K})$)	Thermal Conductivity $\text{W}/(\text{m}\cdot\text{K})$	Coefficient of Linear Expansion
GeTe-Sb ₂ Te ₃ -Sb (Amorphous)	4.9+1.4i	6150	5.49×10^{10}	0.33	0.209×10^3	0.581	1.1×10^{-5}
GeTe-Sb ₂ Te ₃ -Sb (Crystal)	5.7+3.4i	6150	5.49×10^{10}	0.33	0.209×10^3	0.581	1.1×10^{-5}
2 : 1 : 0.5 (mol ratio)							
ZnS-SiO ₂	2.0	3650	7.81×10^{10}	0.2	0.563×10^3	0.657	7.4×10^{-6}
4 : 1 (mol ratio)							
SiO ₂	1.46	2202	7.81×10^{10}	0.2	0.753×10^3	1.313	5.5×10^{-7}
Al Alloy	2.2+7.5i	2750	7.03×10^{10}	0.345	0.892×10^3	0.215×10^3	2.2×10^{-5}
Polycarbonate	1.58	1200	2.26×10^9	0.3	0.126×10^2	0.223	7.0×10^{-5}

Latent Heat (Crystal \leftrightarrow Liquid) $0.682 \times 10^5 \text{ (J/kg)}$
(Amorphous \leftrightarrow Liquid) $0.356 \times 10^5 \text{ (J/kg)}$

Thermal expansion coefficients of the dielectric materials of SiO_2 and ZnS-SiO_2 are 5.5×10^{-7} and 6.1×10^{-6} , respectively. We compared the thermal deformations of the first version disk structure and new disk structure. Figure 8(a), (b) shows the simulation results of the thickness change of the layers by thermal expansion as the temperature distribution simulation. All layers show that the thermal expansion and thickness changes are several sub-nm. These deformations are considered the motivating force of the micro-displacement of the active layer components. Figure 8(a) shows the thickness change of the first version disk structure. Figure 8(b) shows the thickness change of the new disk layer structure. When the SiO_2 layer for the ZnS-SiO_2 layer is introduced to the upper dielectric layer, the thickness change of the upper dielectric layer becomes negligible small, compared the value of ZnS-SiO_2 of $0.04 \text{ nm}^{(1)}$. The new disk structure, which has a SiO_2 upper dielectric layer, produces more than 1,000,000 overwrite cycles. Figure 9 shows the results of one million overwrite cycle test of the disk of SiO_2 upper dielectric layer. The Error bit counts, C/N ratio, Erase ratio and Jitter (window % for 10^{-4} bit error rate) are almost stable through the test.

A conventional laser recording using rather long pulse widths of 10 ns to 60 ns (10^{-9} s) shows large thermal diffusion phenomena in the disk. The pulse means that the irradiation time of a laser spot at a certain point on the rotational disk, and is the time the laser spot takes to go through the point. A short pulse width, such as a femto second (10^{-15} s), will be expected to suppress the thermal diffusion phenomena extremely. The interaction effect of the femto second laser pulse on phase-change films will be discussed later in this paper.

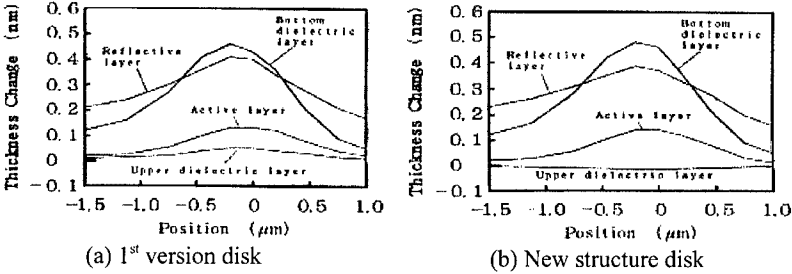


Fig. 8. Distribution of the thickness changes of the disk layers by thermal expansion.
 1st version disk (a): substrate / DL, 160nm / AL, 20nm / DL, 35nm / RL, 130nm
 DL : ZnS-SiO_2 , AL : $\text{GeTe-Sb}_2\text{Te}_3\text{-Sb}$, RL : Al-alloy
 New disk (b): upper DL is replaced by SiO_2 (35nm)
 Horizontal axis : laser passing direction, Zero position : center of the laser spot

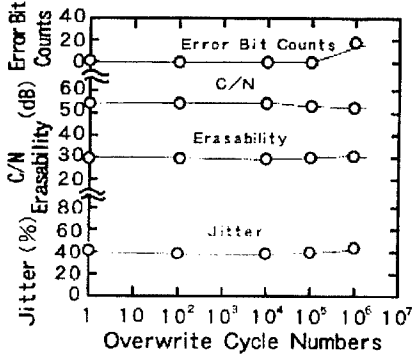


Fig. 9. One million overwrite characteristics of a phase-change optical disk with SiO_2 upper dielectric layer.
 Linear velocity: 11m/sec

3.3 Nitrogen doped GeTe-Sb₂Te₃-Sb active layer

In the phase-change optical disk media development procedure of 1990, we found curious phenomena in the phase-change sample disks. Almost periodically, special disks were sputtered that showed a number of overwrite cycle characteristics almost 10 times larger than the other disks. Our investigation found that these special disks were formed right after the sputtering chamber was cleaned. This means that some adsorbed components in the chamber during the chamber cleaning improved the cycle.

Air is comprised of nitrogen, oxygen, some H₂O, and so on. For the chemical stability of the media, we choose nitrogen as a doping component in the active layer. We control the quantity of nitrogen doping by a gas flow ratio of N₂/Ar into the sputtering chamber. The active layer of doped nitrogen shows an N₂ doping dependency of the optical constant (n, k), an increasing N₂ component, and a decreasing refractive index n and extinction coefficient k. This means that nitrogen forms nitride material of M-N in the active layer. Usually, nitride materials show a high melting temperature, which is believed to indicate suppression of the micro-displacement of the active layer components through the overwriting cycle measurement¹²⁾.

Figure 10 shows the XPS (x-ray photoelectron spectroscopy) measurement results of the N₂-doped phase-change film. The spectral data shows a large peak at the binding energy of 397.2 eV, which corresponds to Ge-N bonding.

After overcoming the cycle issue by key technologies, the phase-change optical disk became a reliable data-recording disk, and its advantage of ROM (read-only memory) disk compatibility it becomes to be multimedia optical disks such as PD, CD-RW and rewritable DVD. We developed a high-density 90 mm diameter phase-change optical disk for an ISO (International Organization for Standardization) standardization proposal in 1995¹³⁾. This disk featured top-level technologies such as a red light laser diode, a large NA (numerical aperture) (NA=0.6) lens and a thin disk substrate (0.6 mm)^{14),15)}. A thin disk substrate is effective for resolving the disk tilt problem during high-density recording. A thin disk substrate of 0.6 mm thickness has lower cross-talk characteristics. These technologies were adapted in the 4.7GB DVD in 1995.

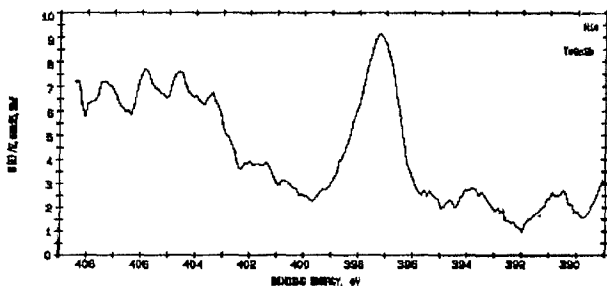


Fig. 10. XPS analysis of GeTe-Sb₂Te₃-Sb film sputtered in N₂/Ar atmosphere. Binding energy of Ge-N : 397.2eV.

4. High density recording technologies of phase-change optical disks

4.1 Density competition between MO disk and Phase-change optical disk

Magneto-optical disks have recently been demonstrated to show the unique capability of MSR (magnetic induced super-resolution)¹⁶⁾, and now feature MAMMOS (magnetic amplifying magneto-optical system)¹⁷⁾ and DWDD (domain wall displacement detection)¹⁸⁾ for high-density recording using both an optical pick-up and a magnetic head. These features provide small-mark detection, high signal output and ISI (inter-symbol interference) free high-density reading methods, respectively. They have a recording density of around 11 Gbit/in² using a conventional optical system (laser wavelength = 650 nm, lens numerical aperture NA = 0.6¹⁹⁾). However they have certain drawbacks, such as a complex disk structure composed of both a recording magnetic layer and a reading magnetic layer, as well as a sensitivity variation caused by ambient temperature during both recording and reading.

The phase-change recording layer has the advantages of a high-signal output and response to a wide wavelength spectrum. Table 2 shows the wavelength dependency of the complex refractive index of the phase-change material film in the amorphous and crystalline state, which shows that the phase-change film has a wide wavelength response.

Three main approaches have been proposed to increase the recording density of phase-change optical disks. The first is to combine a short wavelength laser with a large NA lens, recently DVR technology is proposed, which is applying larger numerical aperture lens of NA=0.85 and the new disk structure of thin cover-layer²⁰⁾, the thickness is 0.1mm for 0.6mm disk substrate. The other is SIL(solid immersion lens) and Near-Field technology, which is effective for surface recording. The recording density increases to double that of a conventional DVD and the blue laser increases the capacity to 23 GB²⁰⁾.

The second is dual layer recording, creating volumetric rather than two-dimensional surface recording. The dual-layer phase-change optical disk has a density of 6.4 Gbit/in² using a conventional optical system (laser wavelength = 650 nm, lens numerical aperture NA = 0.6²¹⁾). This technology features enhanced density and compatibility with DVD pick-up. The third is the possibility of multi-level recording on phase-change optical disks.

4.2 Blue laser Dual-layer phase-change optical disk

Dual-layer recording is another form of volumetric technology. The first such commercial product was a dual-layer DVD for a DVD 8.5 GB ROM disk for a cinema title.

Rewritable 8.5 GB phase-change dual-layer experimental results were announced in 1998¹⁹⁾. Figure 11 shows the blue laser 27 GB basic dual-layer disk structure²²⁾.

Table 2 Wavelength dependency of complex refractive index ($N = n + ik$) of phase-change material GeTe-Sb₂Te₃-Sb film.

Wavelength	Refractive index	
	Amorphous	Crystal
830nm	4.61+1.05i	5.67+3.01i
780nm	4.47+1.40i	5.07+3.42i
650nm	4.21+1.89i	4.56+4.23i
430nm	3.08+2.51i	2.21+3.77i
405nm	2.90+2.51i	2.03+3.58i

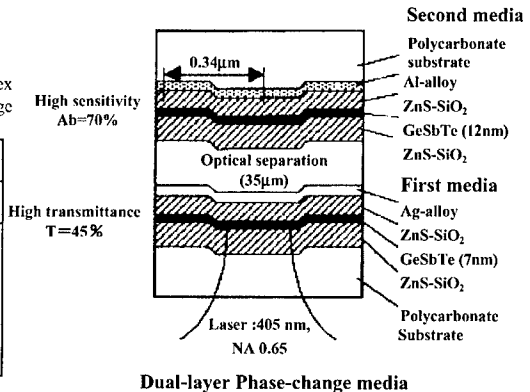


Fig. 11. Cross-sectional view of the Blue laser Dual-layer rewritable Phase-change optical disk.

The first medium has a high transmission characteristic of 45% in the crystalline state with a recording reflectivity difference of 7%. The second medium has a high reflectivity-difference signal output of 24% combined with a high sensitivity characteristic. As a result, the signal output of the second medium becomes $24 \times 0.5 \times 0.5 = 6\%$ by the absorption of the first medium, which closely matches the value of the first medium. The blue laser, DVD compatible optical pick-up, NA = 0.6 and 0.6 mm substrate method increases the capacity to 27 GB.

4.3 MRWM (mark radial width modulation) concept of multi-level recording

Multi-level recording was first announced in a phase-change electrical switching memory (Ovonic memory) device in 1997 that had 16 switching levels²³⁾.

The phase-change optical recording layer has a large reflection-difference characteristic between the crystalline state (around $R_{cry} = 30\%$) and the amorphous state (around $R = 7\%$) of the same order as in the CD-ROM pit signal output. M. P. O'Neill demonstrated an 8-level phase-change recording technology at ODS2000 and 2 GB of CD-RW capacity capability²⁴⁾.

We propose to subdivide this large signal output into multi-level (ML) signals on a phase-change optical disk using the MRWM (mark radial width modulation) method²⁵⁾.

Figure 12 shows the TEM observation of MRWM recording marks on a phase-change optical disk. The mark radial widths are 200 nm, 400 nm and 600 nm for level 1, level 2 and level 3, respectively. The idea of MRWM recording is to assign a specific laser pulse width and power level to a specific mark level. The assigned laser pulse widths are 114 ns, 84 ns and 46 ns, and the assigned laser power is 6 mW for Level 1, 7 mW for Level 2 and 11 mW for Level 3. Figure 13 shows the C/N (carrier-to-noise) and amplitude of the MRWM recording method with various pulse widths and power conditions. The C/N value is 50.1 dB, 57.5 dB and 61.5 dB for the Level 1, Level 2 and Level 3 marks, respectively. The minimum mark length is defined by the beam factor of the optical path and is controlled by the assigned power level and the pulse width.

5. Combination technology of high-density recording

The density of one side of the 4.7 GB version rewritable DVD holds 3.4 Gbit/in². A dual-layer phase-change rewritable disk whose capacity is 8.5 GB has an effective density on one side of 6.4 Gbit/in², increasing the density 1.9 times. By introducing the magnification factor of the multi-level recording of $M = 4$ ($\times 1.76$) to $M = 8$, the recording density will further increase approximately 2 to 3 times.

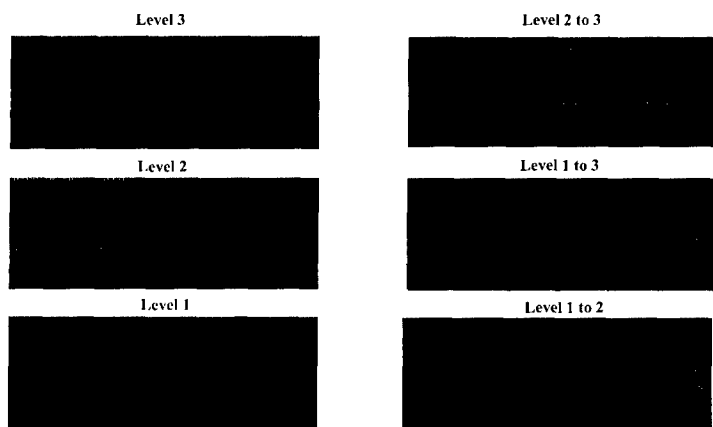


Fig. 12. TEM observation of MRWM recording marks (Track width: 600nm)
Each marks, Level 1, 2, 3 and the set recording marks (1 to 2), (1 to 3), (2 to 3)

Another density increasing strategy is to apply a large numerical aperture lens of NA = 0.85 and a 0.1 mm-thin overcoat layer for 0.6mm substrate. The recording density will increase to double that of an equivalent DVD due to the factor of 0.85/ 0.60. The blue-violet laser wavelength of 405 nm will increase the density to approximately 2.6 times that of the 650 nm laser system. The recording density is predicted to be 63.2 Gbit/in² and the capacity will rise to 87GB/120mm/side. By magnifying the multi-level recording from M = 4 to M = 8, the density is expected to increase to more than 100 Gbit/ in².

J. Tominaga has announced a phase-change Super-RENS(super-resolution near-field structure) recording technology, which achieves 13 Gbit/in² using the conventional optical system (laser wavelength = 640 nm, lens numerical aperture NA = 0.6²⁶). The Super-RENS effect can be combined with the above technologies, resulting in a potential density increase of approximately four times to achieve 250 Gbit/in² in the future.

Near-Field recording technology is now actively developing, applying SIL technology on phase-change optical disks with GaN blue laser. K. Kishima demonstrated a 40 Gbit/in² recording density at ODS2000²⁷. Figure 14 shows the area density expansion of the phase-change optical disk with the combination of high density recording technologies.

There are two strategies for next generation high density phase-change optical disk, the one is DVD compatible strategy which keeps the lens numerical aperture (NA=0.6) optical pick-up and the thin substrate (t=0.6mm) DVD disk structure and combines dual-layer technology and multi-level technologies so on. The other is new disk strategy different from DVD and is introducing larger numerical aperture (NA=0.85) optical pick-up and thin cover-layer (t=0.1mm) disk structure which is not compatible with DVD but can be introduced the technologies, such as dual-layer or multi level recording.

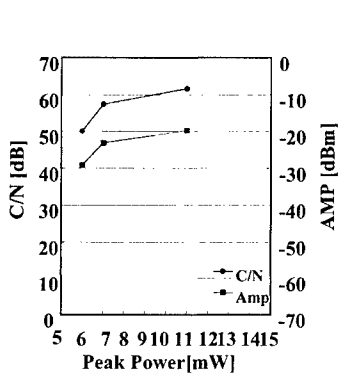


Fig. 13. Pulse width and Power assigned MRWM recording characteristics of Phase-change optical disk.

Level 1(113.3ns, 6mW)
 Level 2 (83.1ns, 7mW)
 Level 3 (45.3ns, 11mW)
 Signal: f=6.6MHz, v=8.2m/s

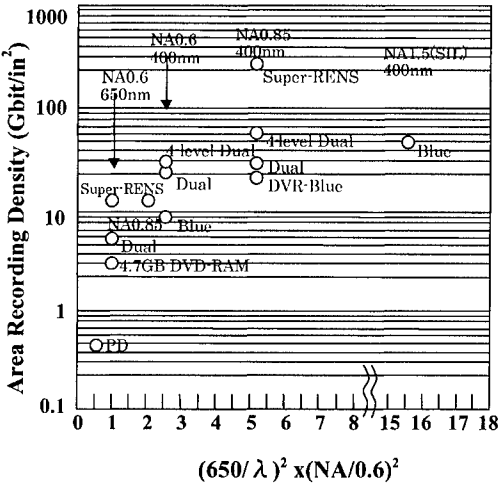


Fig. 14. Areal recording density growth of Phase-change optical disk.

PD (λ =780nm, NA=0.5), 4.7GB DVD-RAM
 $(\lambda$ =650nm, NA=0.6), 8.5GB Dual-layer disk,
 Blue wavelength 27GB Dual -layer disk, DVR,
 SIL (NA=1.5)
 Multi-level recording

The phase-change media sample structure is a polycarbonate substrate/ZnS-SiO₂ 155nm/GeSbTe 24nm/ZnS-SiO₂ 45nm/Air without a reflection layer, making it easy to observe the mark formations with the optical microscope. The structure of the conventional laser recording media has a reflection layer on the above sample and the recording is from the substrate side.

6.3 Femto second laser recording mark and discussions

Figure 16 (a) shows the TEM observation of the mark of the conventional several-tens nanosecond long pulse-width laser exposure (50 ns) on the rotational disk and (b) shows the mark formed by the femto second laser exposure (120 fs). The lens numerical aperture was $NA = 0.5$, the laser wavelength was $\lambda = 780$ nm, and the recording power was 11 mW. The marks are amorphous and surrounded by a large crystalline band edge.

The most significant difference between the mark formed by the femto second pulse laser and the marks formed by the conventional several-tens nanosecond pulse laser is the mark edge figure. The conventional pulse forms intermediate space between the amorphous mark and the crystalline background dead space of 160 nm. The femto second pulse forms an amorphous mark without the crystalline edge band. The intermediate space is a large-grain size crystalline state that is formed by the recording laser spot when melted and re-crystallized in the cooling step. The recording energy density of the femto second mark of the phase-change media was $1\text{ nJ}/\mu\text{m}^2$, and the conventional disk recording energy density was approximately $2\text{ nJ}/\mu\text{m}^2$.

The conventional laser disk recording process has three stages. The first is a laser spot exposure, the second is heat diffusion outside of the laser spot, and the third is cooling after the laser spot passes away from the portion. The laser exposure time is from 10 ns to 100 ns, depending on the laser spot diameter and the disk rotational speed. The conventional laser recording method appears in the second and third process, and comes from the thermal diffusion process. The mark dead space out of the laser spot and the variation of the mark position are caused by the thermal diffusion from a rather long pulse-laser irradiation.

The femto second laser pulse irradiation is quite different from the conventional laser recording because the thermal diffusion is limited in the fs to ps order, the mark is accurately formed in the laser spot, and the variation of the mark position is negligible without thermal diffusion variation. The short pulse width of 120 fs recording on the phase-change media indicates a high-recording data rate capability of more than 1 Tbit/s.

Overwrite process needs ultra high speed crystallizing characteristics and now investigations start for this new material area.

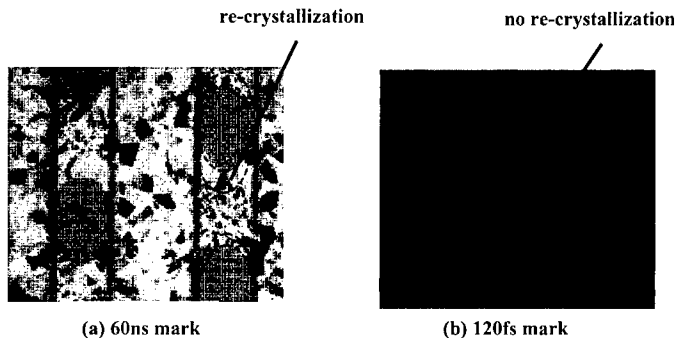


Fig. 16. TEM observation of the amorphous marks on phase-change optical disk.

(a) Conventional laser recording : $\lambda = 780\text{ nm}$, $NA = 0.5$, $t = 60\text{ ns}$, (with a reflection layer)

(b) Femto second pulse laser recording : $\lambda = 800\text{ nm}$, $NA = 0.95$, $t = 120\text{ fs}$, (without a reflection layer)

7. Conclusion

Key technologies obtained by materials research and disk structure development have achieved multimedia rewritable 4.7 GB DVD products. The application of blue laser light and volumetric (dual layer) recording have the potential to increase the recording density to

20 Gbit/in² with an NA = 0.6 lens. The phase-change optical disk has multi-level high-density recording capability. A multi-level recording method and an NA of 0.85 in combination with other technologies indicates the potential for the recording density of the phase-change optical disk to exceed 100 Gbits/in².

Ultra-short pulse femto second laser recording on phase-change media is one candidate for a breakthrough of the laser thermal recording limit.

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